The Abdus Salam
International Centre for Theoretical Physics

The International Union of Geodesy and Geophysics

United Nations
Educational, Scientific and
Cultural Organization

IAEA
International Atomic Energy Agency

2339-2

Workshop on Atmospheric Deposition: Processesand Environmental Impacts

21 - 25 May 2012

Radionuclide Deposition: The Fukushima Dai-ichi Nuclear Power Facility Incident

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Radionuclide Deposition: The Fukushima Dai-ichi Nuclear Power Facility Incident

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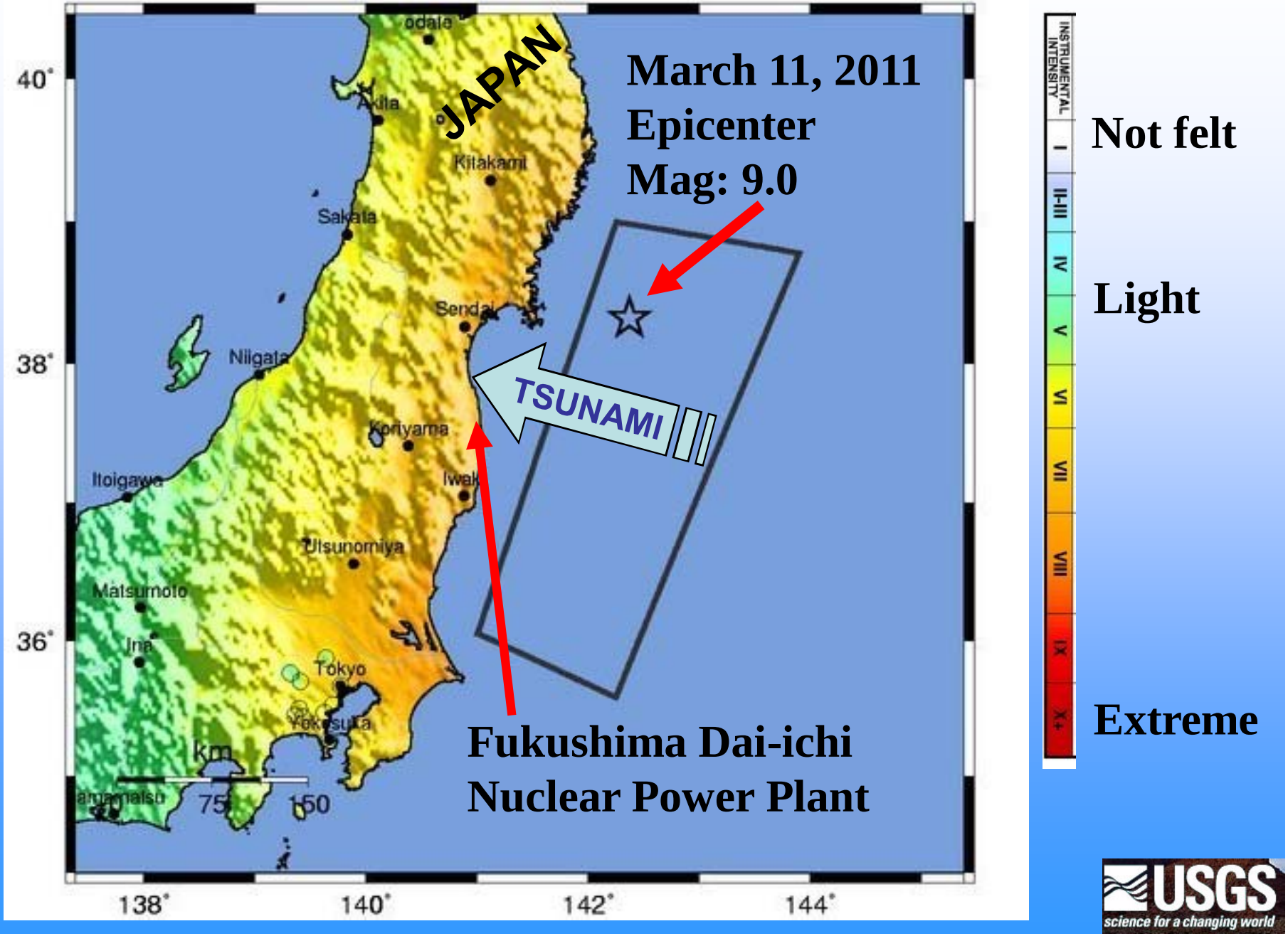
Timothy M. Debey[1]
Christopher M.B. Lehmann[2]
Mark A. Nilles[1]

1 USGS science for a changing world

2 National Atmospheric Deposition Program

USGS SHAKEMAP, E. COAST JAPAN, MAR. 11, 2011



Fukushima Dai-ichi Nuclear Power Plant, Near Sendai, Japan

Timeline

March 12, 2011 – Fukushima Dai-ichi Release(s)

March 14, 2011 – NADP and USGS begin preparation of Sampling and Analysis Plans.

- NADP begins saving filters and water samples.

March 15, 2011

- NADP/USGS contacted USEPA to offer samples for analysis. Sent 5 samples on March 28.

March 16, 2011

- NADP/USGS contacted DHS to offer samples for analysis. No samples sent.

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Timeline

March 25, 2011 - USGS Reactor Facility Group (RFG) started analysis of filters.

April 15, 2011 - USGS RFG completes filter analyses

April 18, 2011 - USGS RFG begins water analyses

July 8, 2011 - USGS RFG completes water analyses

August 26, 2011 - Initiated ES&T article and Open-File Report (OFR)

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Timeline

September 2011 - ES&T article and OFR in review

October 18, 2011 - USGS Director's approval for OFR

October 25, 2011 - Presentation of preliminary results at 2011 NADP Technical Symposium, Providence, RI, USA

January 2012 - USGS Director approved ES&T article

February 2012 - ES&T article and OFR Published

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Objectives

1. Evaluate NADP NTN and MDN capabilities for monitoring radionuclide activities in precipitation.

2. Evaluate NADP/USGS capabilities to monitor unexpected atmospheric events.

3. Offer NADP support to agencies responsible for monitoring radioactive fallout – USEPA, DOE, DHS, Environment Canada.

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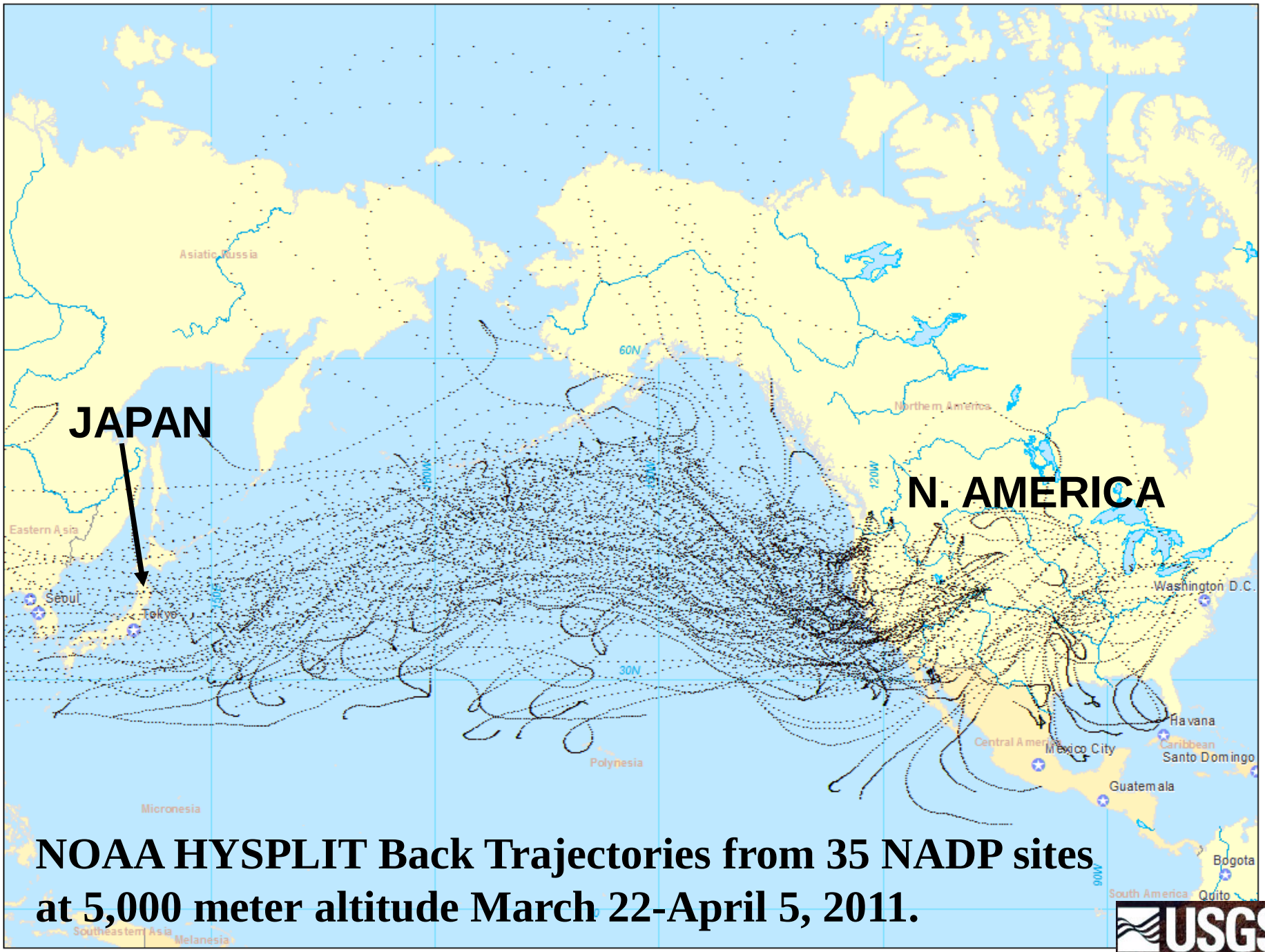

NOAA HYSPLIT Back Trajectories from 35 NADP sites at 5,000 meter altitude March 22-April 5, 2011.

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CO98 and CO89,
Rocky Mountain National Park
3,231 meters altitude
ETI Noah-IV raingages


CO98 and CO89, Rocky Mountain National Park
3,231 meters altitude
ETI Noah-IV raingages


Loch Vale NADP Site: April 19, 2011
After
Collectors
Gages

Sample Processing & Filtration

- Polyethylene buckets and bottles
- Polyethersulfone filters

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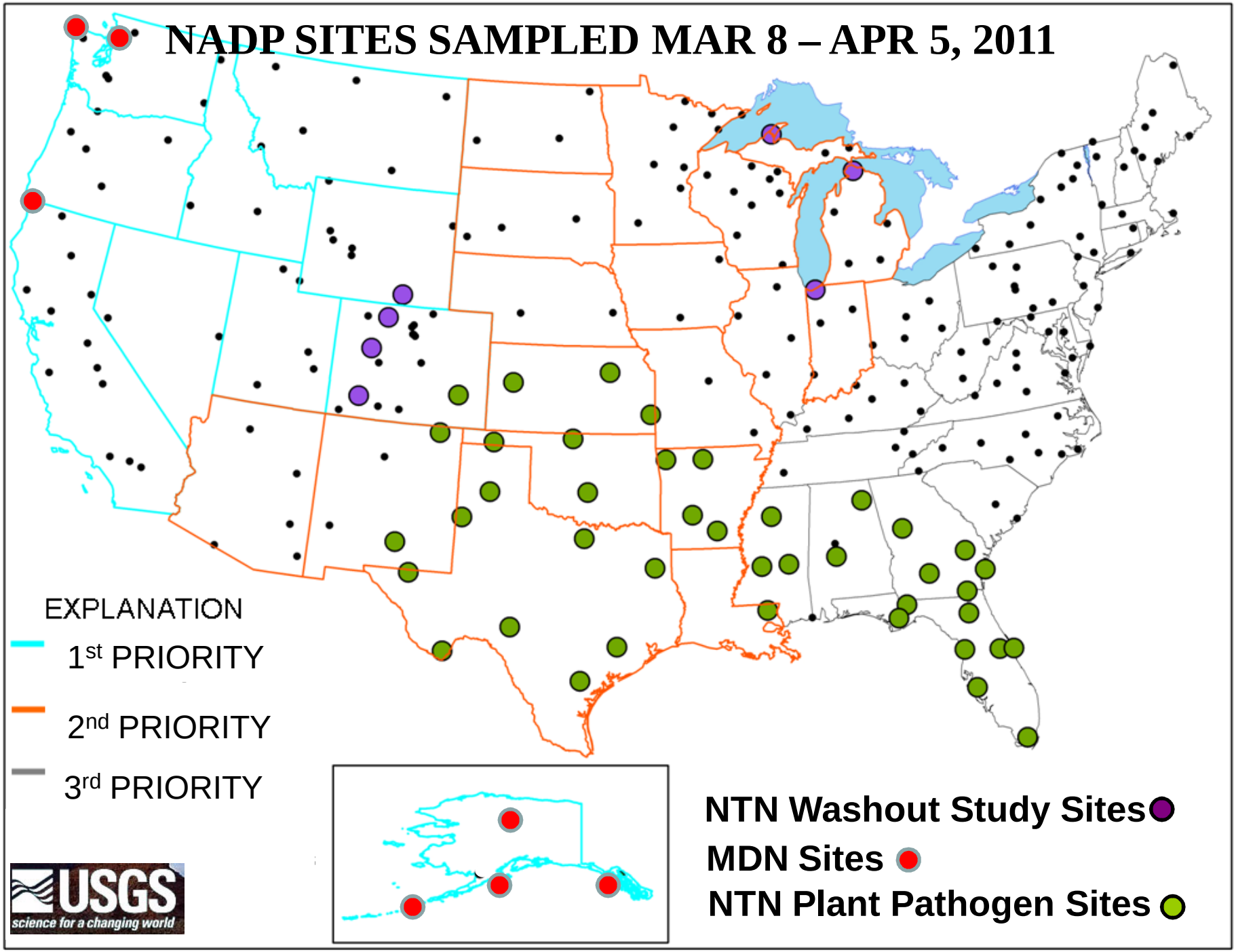
NADP SITES SAMPLED MAR 8 – APR 5, 2011



Filters Sorted by Priority Regions

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Phase 1: 47 mm Diameter Filters

Example of NTN Filter Sample

RESULTS: NO FISSION PRODUCTS DETECTED ON 280 FILTERS.

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WET DEPOSITION SAMPLES
NOT FILTERED
ACIDIFIED, pH < 2.0

Phase 2: Whole-water precipitation samples

First 250 ml, for normal NTN operations

Available for gamma spectrometry

(N = 160 sites)

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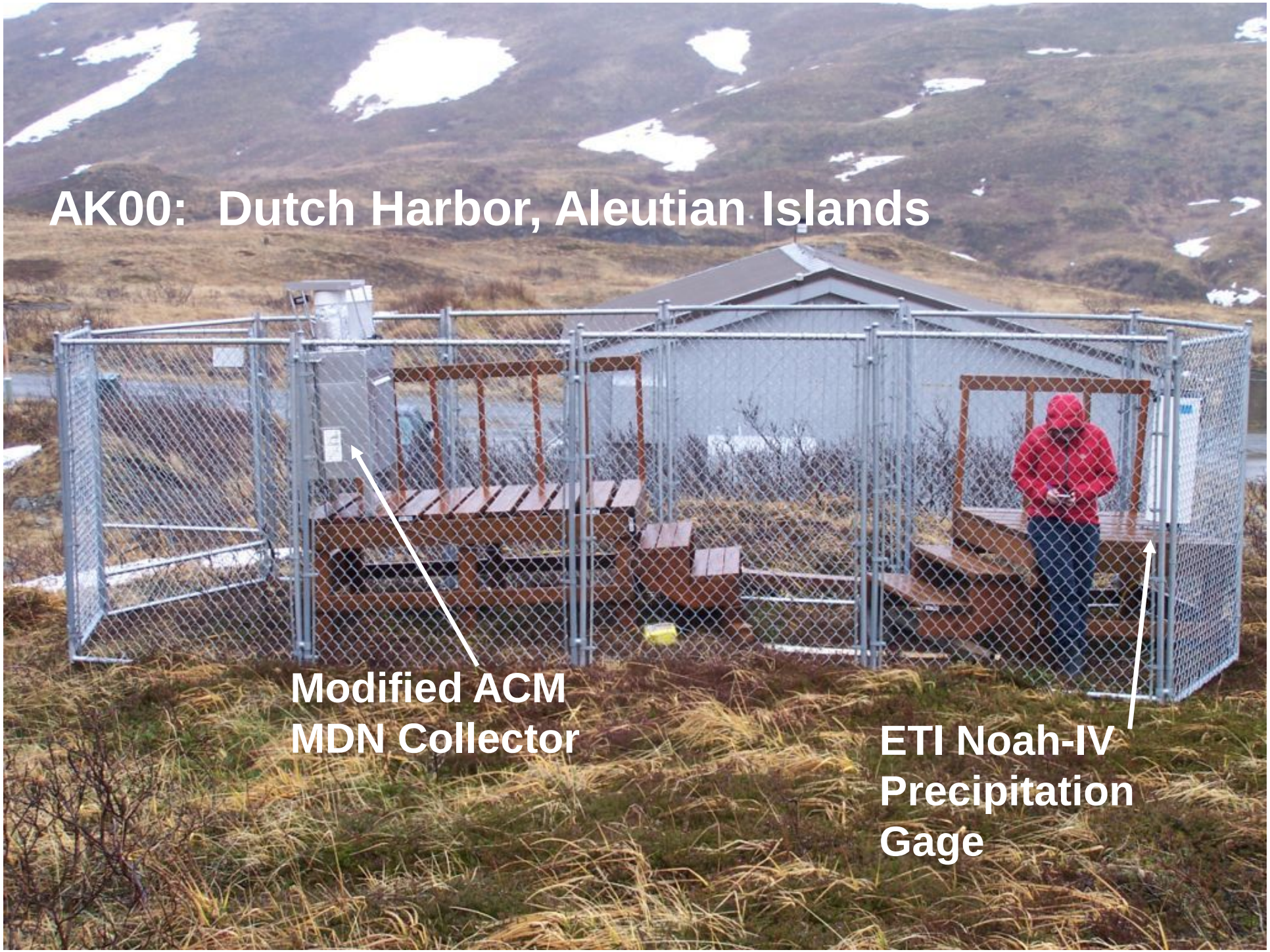
AK00: Dutch Harbor, Aleutian Islands
Modified ACM
MDN Collector
ETI Noah-IV
Precipitation
Gage

Phase 2: Whole-water precipitation samples

N = 16 MDN samples analyzed

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SAMPLES

1. Analyzed 280 NTN filters.

2. Weighed, acidified, composited and analyzed 160 NTN & 16 MDN samples.

3. QA/QC – 8 Blanks and 4 replicates.

4. Gama Spectrometry by
   USGS National Reactor Facility,
   Denver, Colorado

QUALITY ASSURANCE

1. 4 BLANK FILTERS – NO FISSION PRODUCTS

2. 4 BLANK WATER SAMPLES – NO FISSION PRODUCTS

3. SAMPLES FROM WEEK MAR 8-15 ANALYZED FOR SITES WITH DETECTED ^{137}Cs – NO FISSION PRODUCTS

4. REPLICATE SAMPLES FROM CO-LOCATED SITES
   MA01 / 01MA @ CAPE COD NSS – BOTH NO FP
   CA50 / 50CA @ SAGE HEN CREEK FS – BOTH NO FP
   CO98 / CO89 @ ROCKY MTN NP
   – BOTH WITH FISSION PRODUCTS!

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GAMMA SPECTROMETRY

1. 2 DETECTORS AVAILABLE, 16% AND 40% EFFICIENT
2. DETECTOR EFFICIENCY CHECK ^{152}Eu SOURCE
3. RANGE: 122 keV – 1.528 MeV
4. CALIBRATION: +/- 1 keV
5. FILTER CALIBRATION: 0.5 μC ^{60}Co, 0.055 μC ^{137}Cs
6. WATER CALIBRATION: 1 μC ^{152}Eu in 500 mL & 1,000 mL MERINELLI BEAKER GEOMETRY STANDARDS
7. COUNT TIMES: 6 HOURS STANDARD,
   UP TO 24 HOURS FOR RERUNS
8. ^{131}I ACTIVITIES ADJUSTED FOR DECAY FROM TIME OF SAMPLE ANALYSIS TO LAST TIMES COLLECTORS OPEN DURING PRECIPITATION PER RAIN GAGE.
9. ^{134}Cs ACTIVITIES MANUALLY ESTIMATED.

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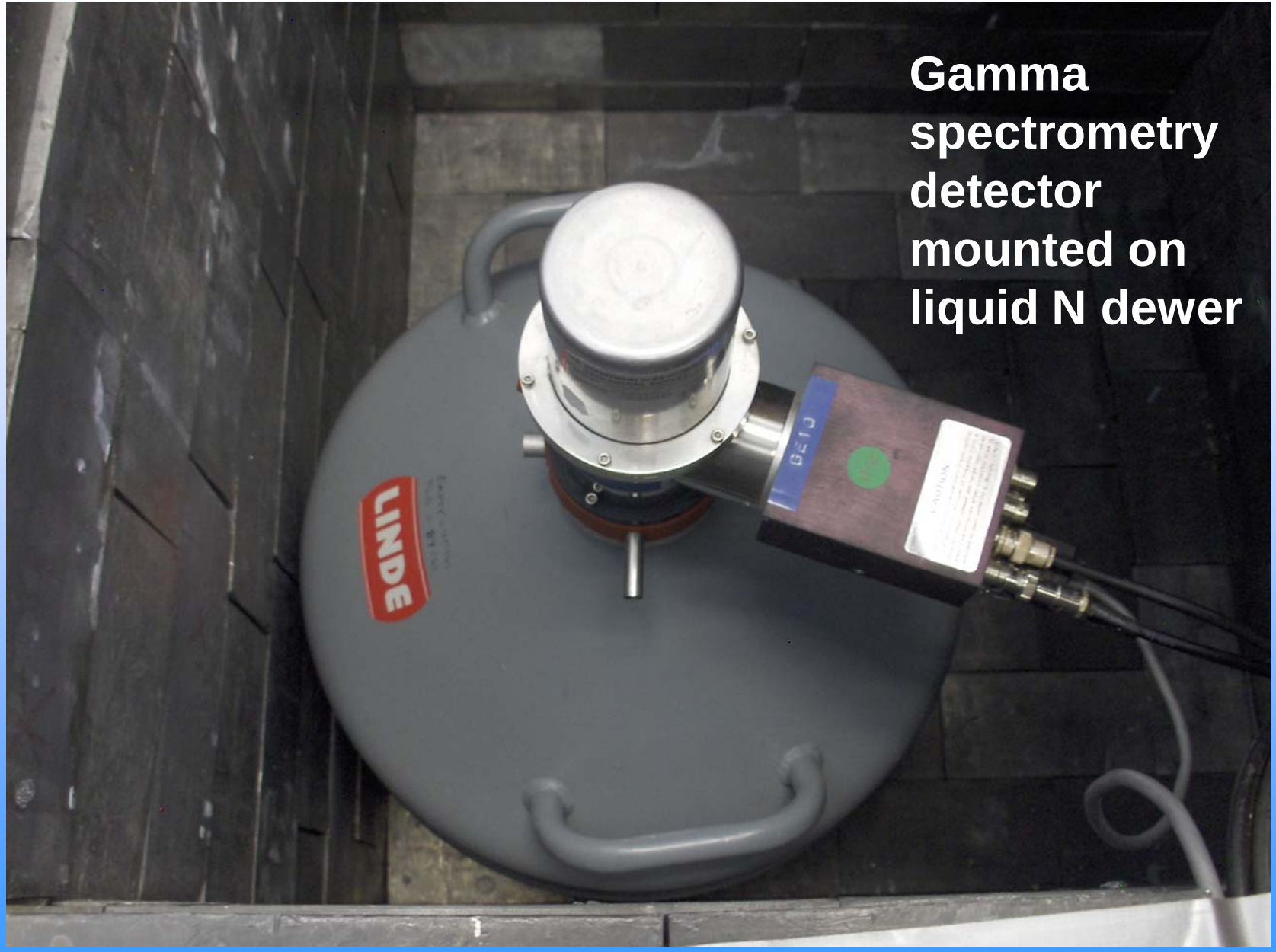
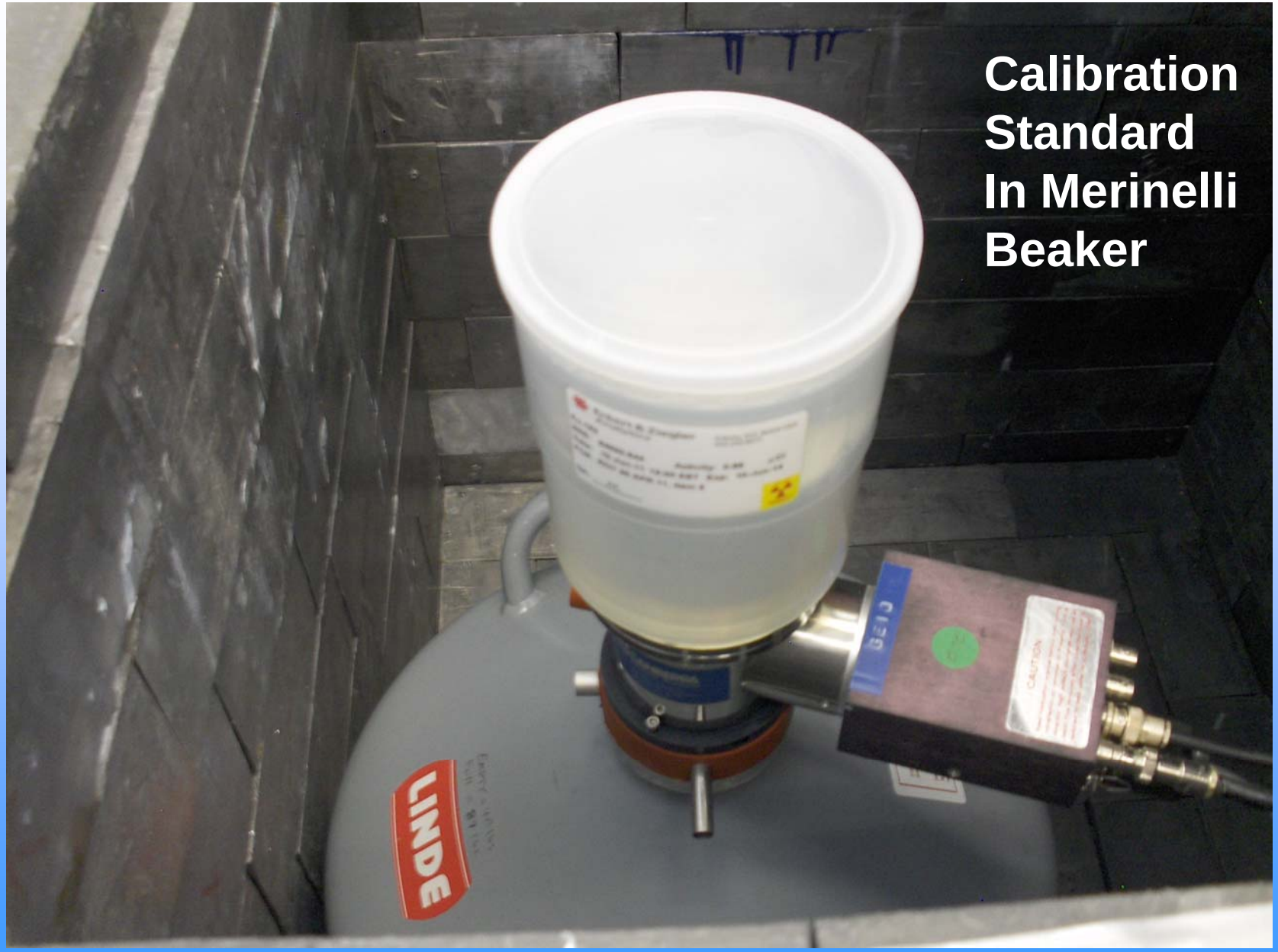
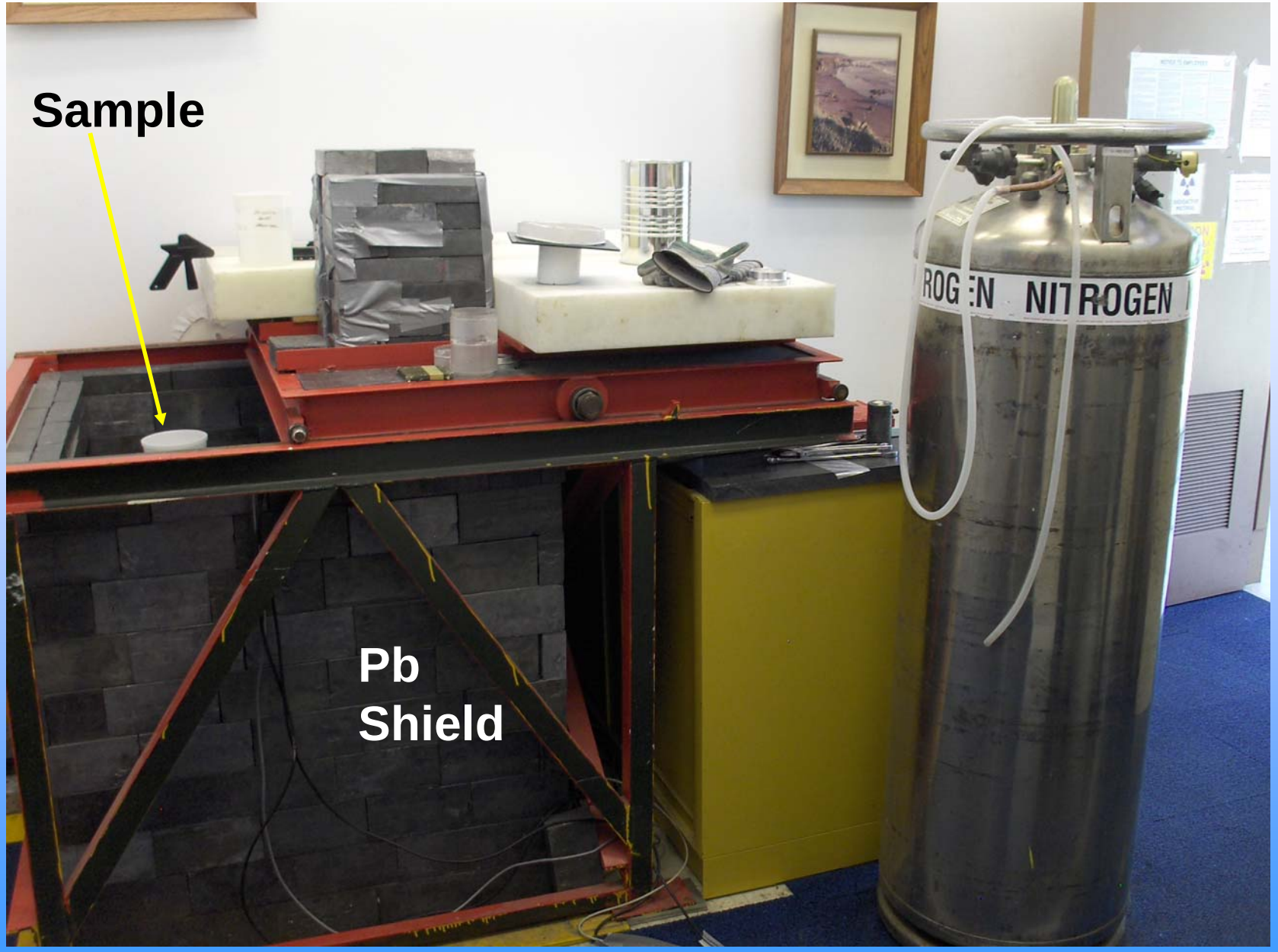
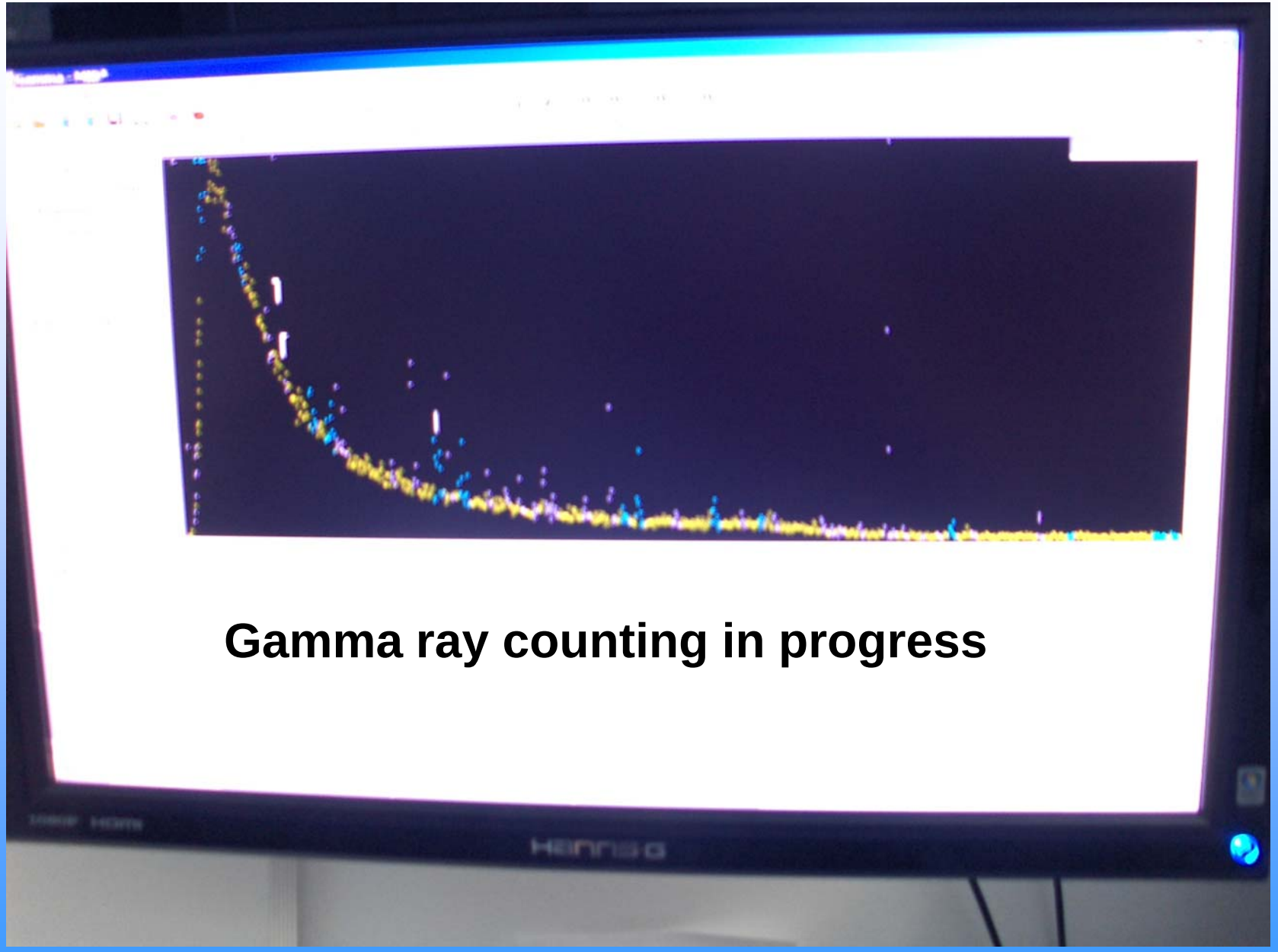

Gamma spectrometry detector mounted on liquid N dewer
LINDE


Calibration
Standard
In Merinelli
Beaker
LINDE


Sample
Pb
Shield
ROGEN NITROGEN



Gamma ray counting in progress

ACTIVITIES vs DEPOSITION (FLUX)

Gamma Spectrometry values in activity units
= picocuries per liter (pCi/L).

Deposition calculated using raingage depths in
= Becquerels per square meter (Bq/m^2)

Conversion Factors: 0.037 Bq / pCi
1 Liter = 1 mm depth / m^2

Deposition, (Bq/m^2)
= ACTIVITY (pCi/L) x PPT DEPTH (mm) x 0.037

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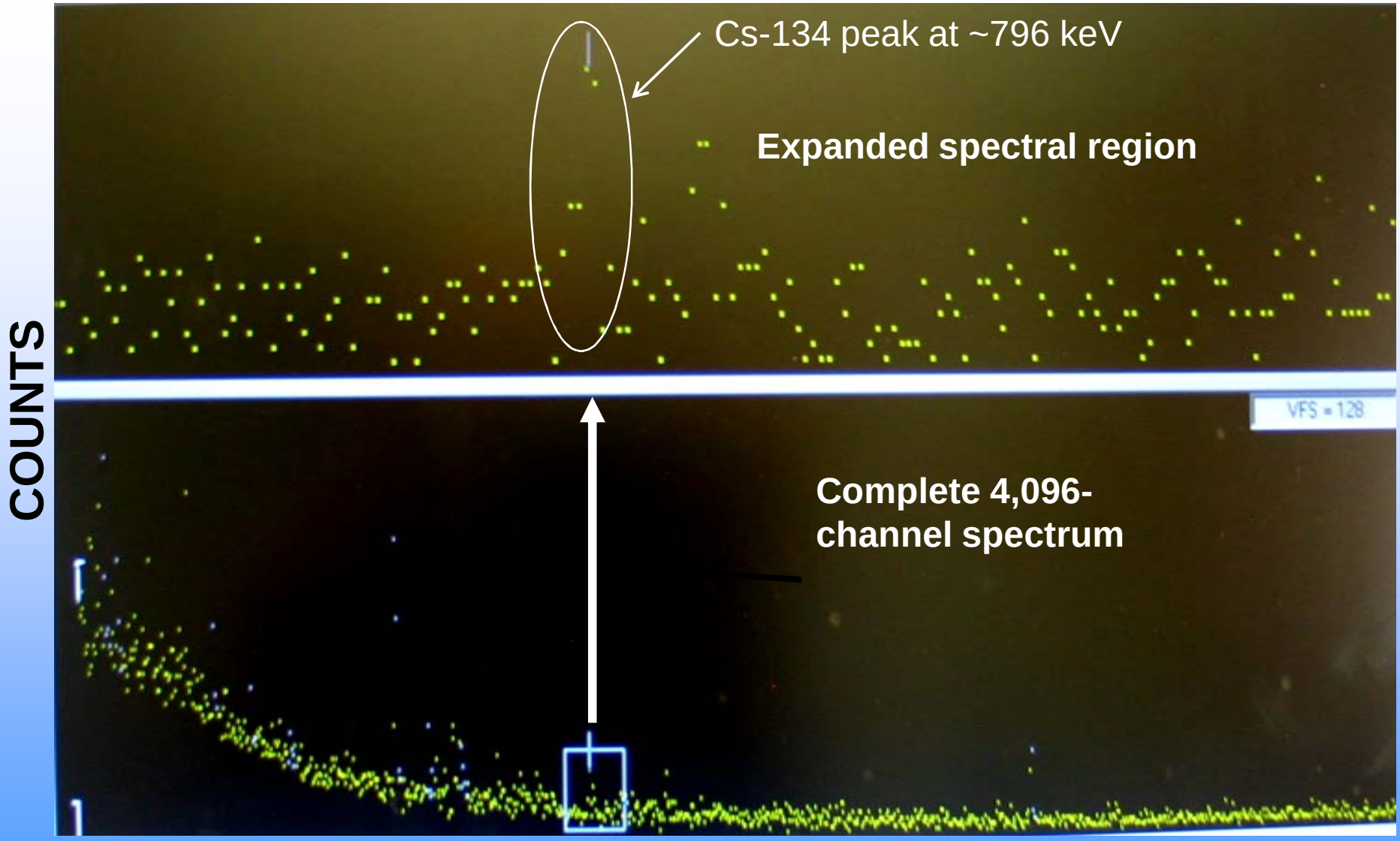
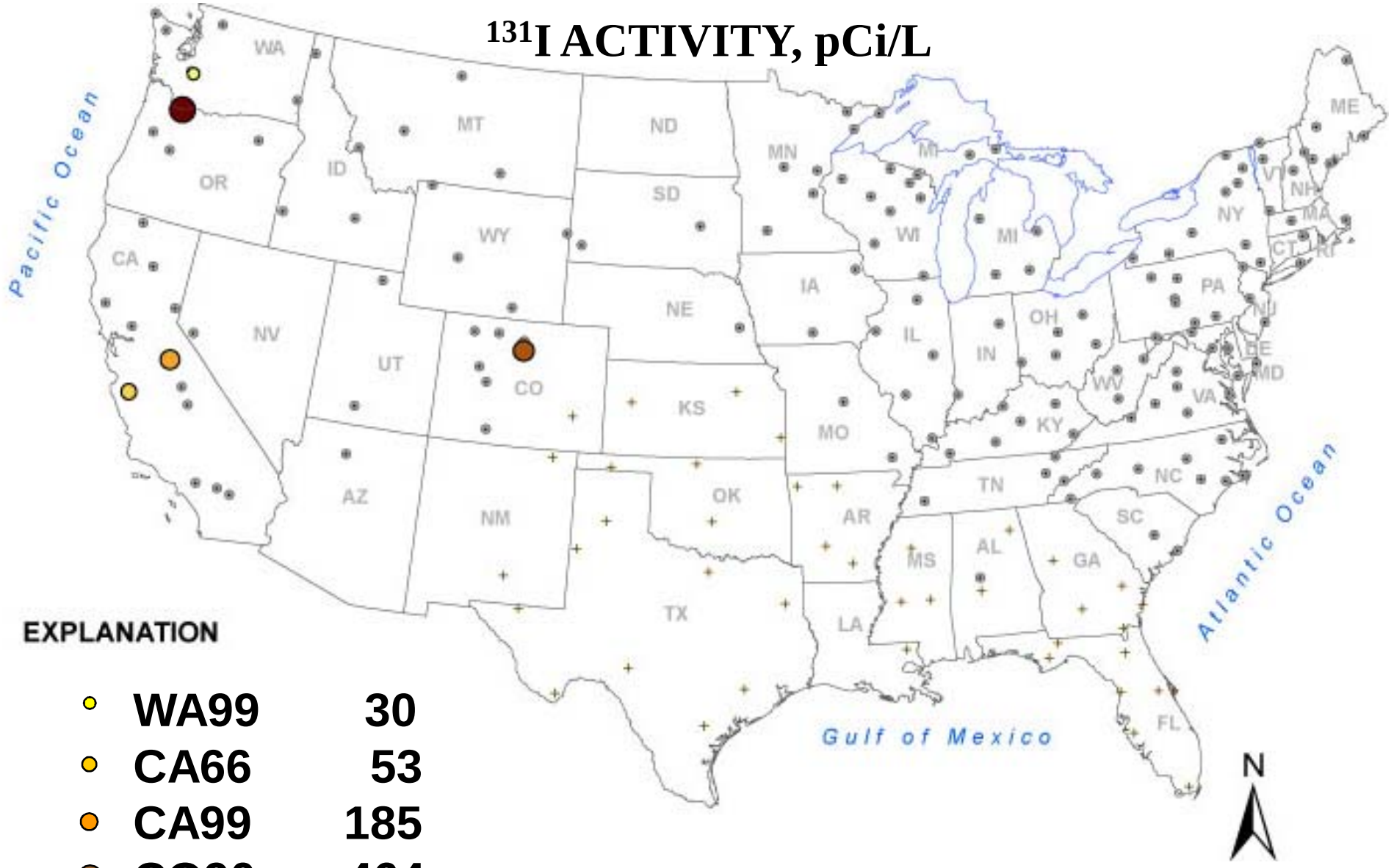


GAMMA RAY EMISSION ENERGY,
IN THOUSAND ELECTRON VOLTS (keV)

^{131}I ACTIVITY, pCi/L



EXPLANATION

- **WA99 30**
- **CA66 53**
- **CA99 185**
- **CO90 464**
- **WA98 1,090**
- **+ Plant Pathogen Study**
- **○ Not detected**

+

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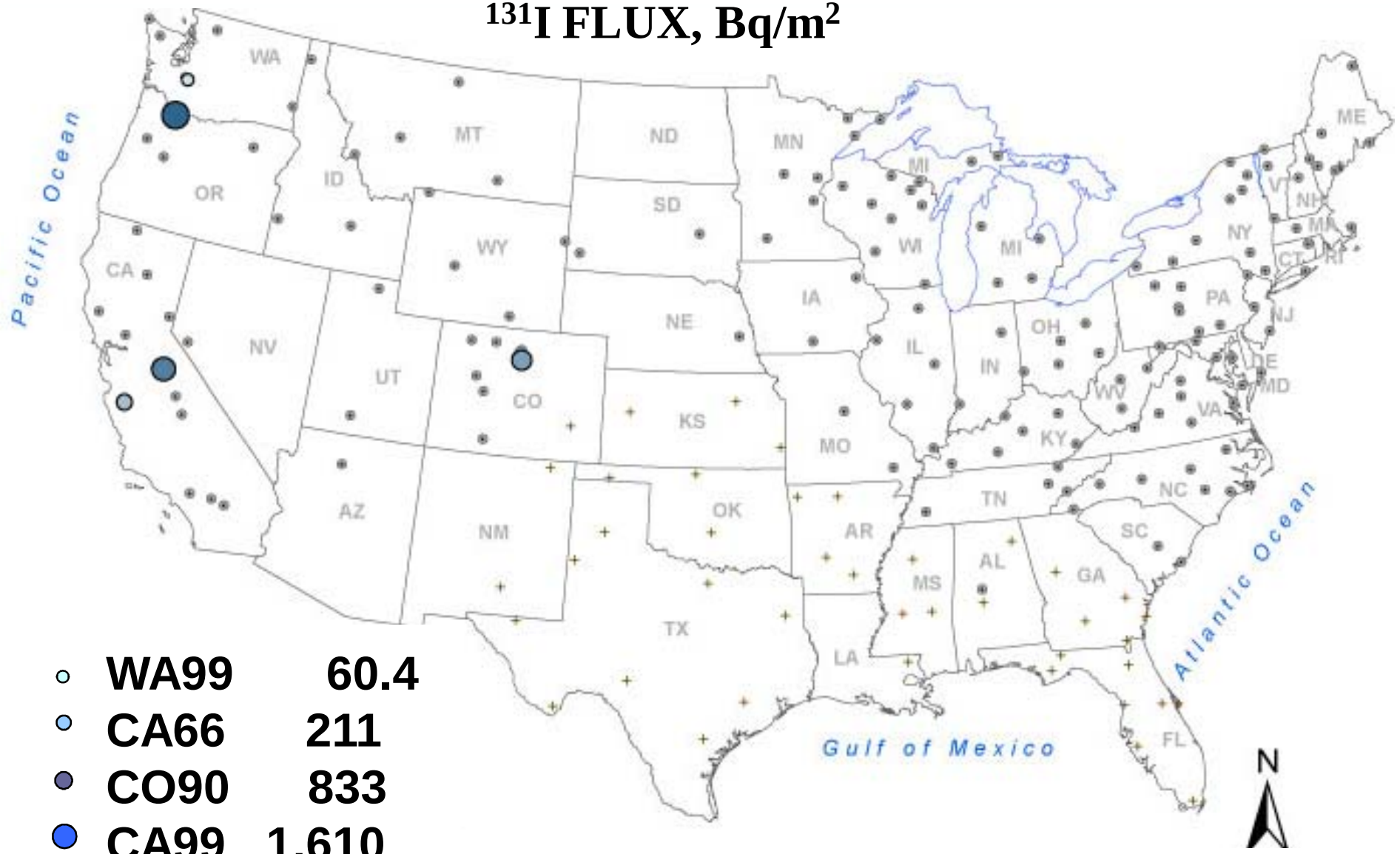
^{131}I FLUX, Bq/m^2



- WA99 60.4
- CA66 211
- CO90 833
- CA99 1,610
- WA98 5,100
- \+ Plant Pathogen Study
- Not detected

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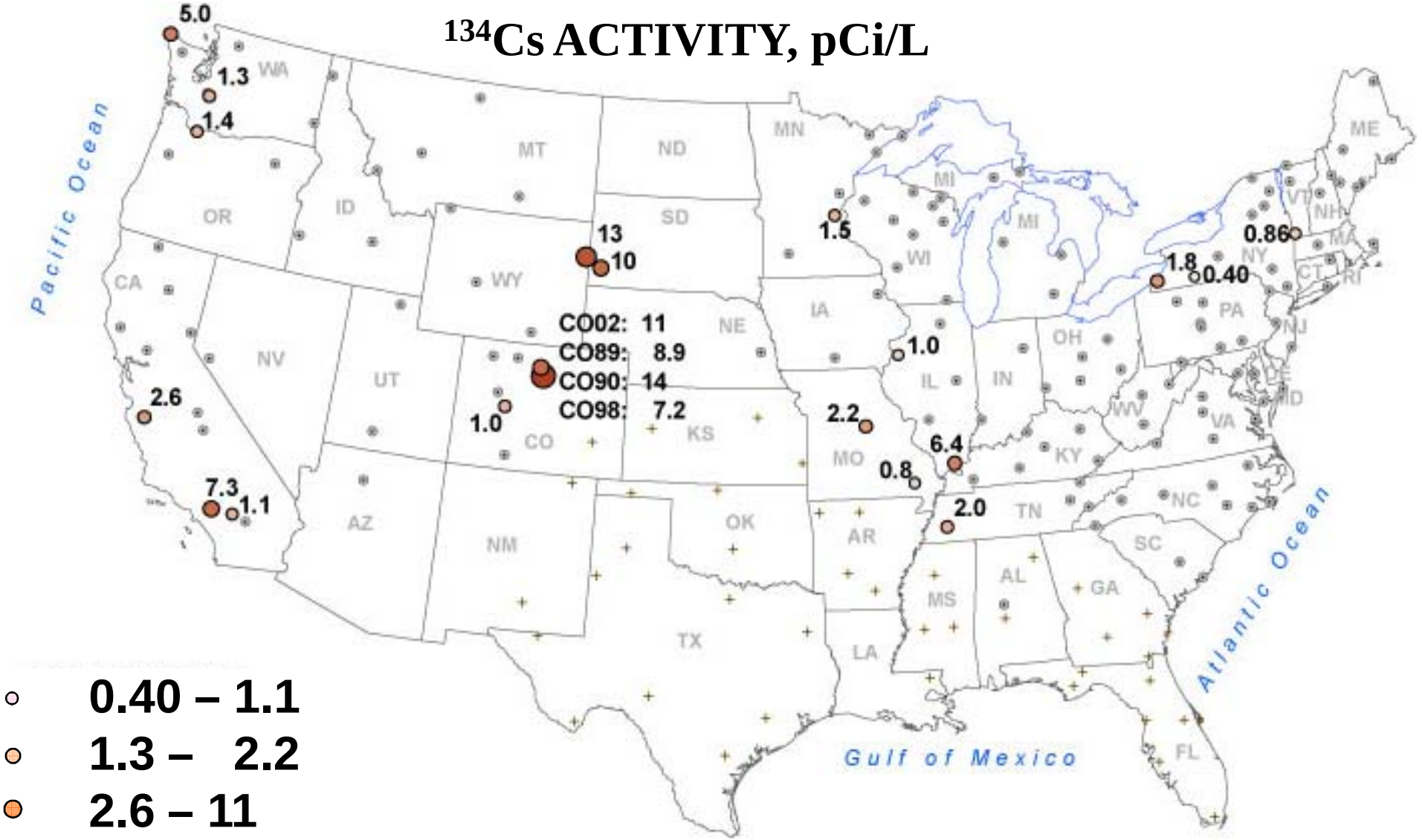
^{134}Cs ACTIVITY, pCi/L



- ○ 0.40 – 1.1
- ○ 1.3 – 2.2
- ○ 2.6 – 11
- ● 11 – 13
- ● 14
- \+ Plant Pathogen Study
- ○ Not detected

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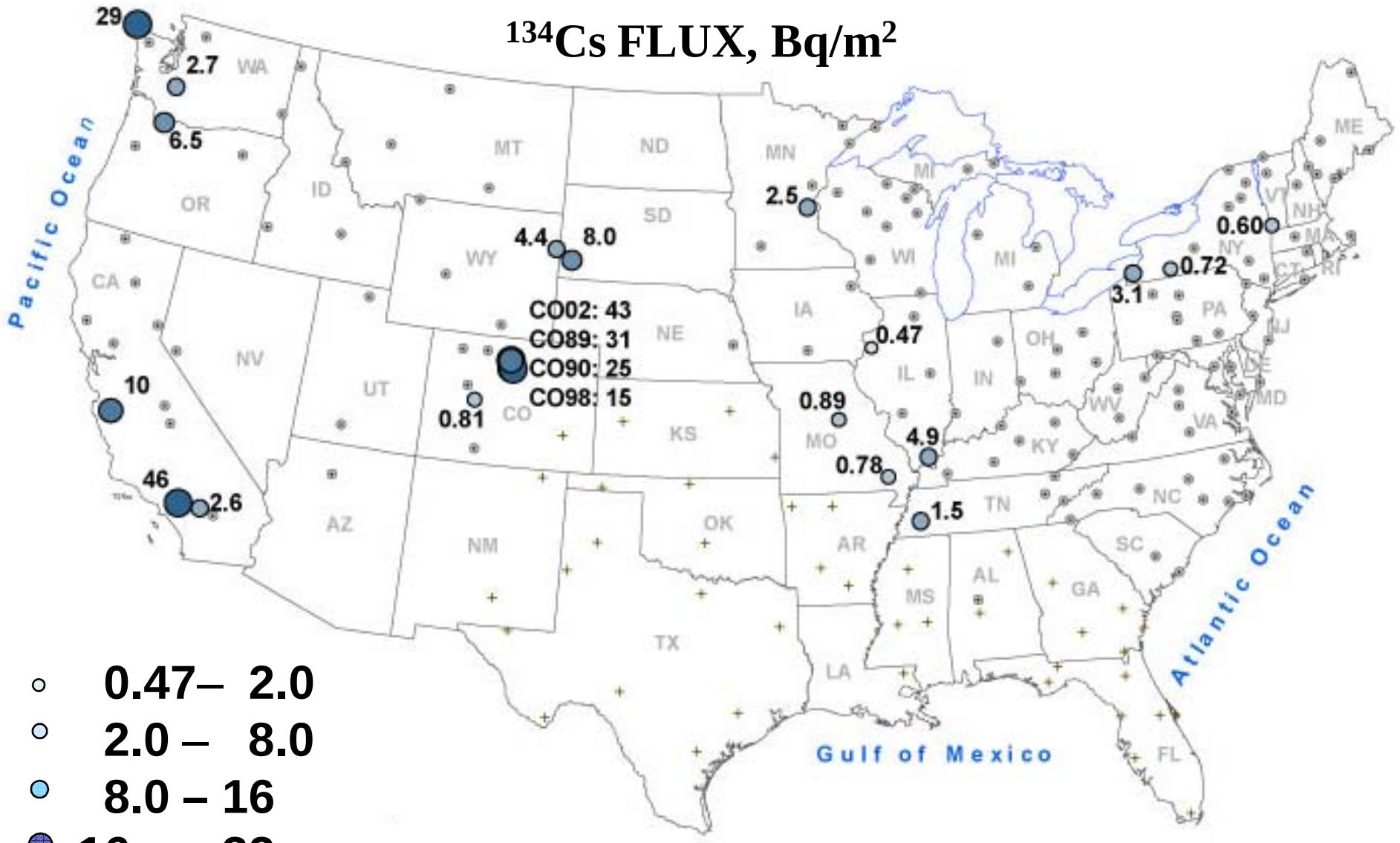
^{134}Cs FLUX, Bq/m^2



- ◦ 0.47– 2.0
- ◦ 2.0 – 8.0
- ◦ 8.0 – 16
- ● 16 – 32
- ● 32 – 46
- \+ Plant Pathogen Study
- ◦ Not detected

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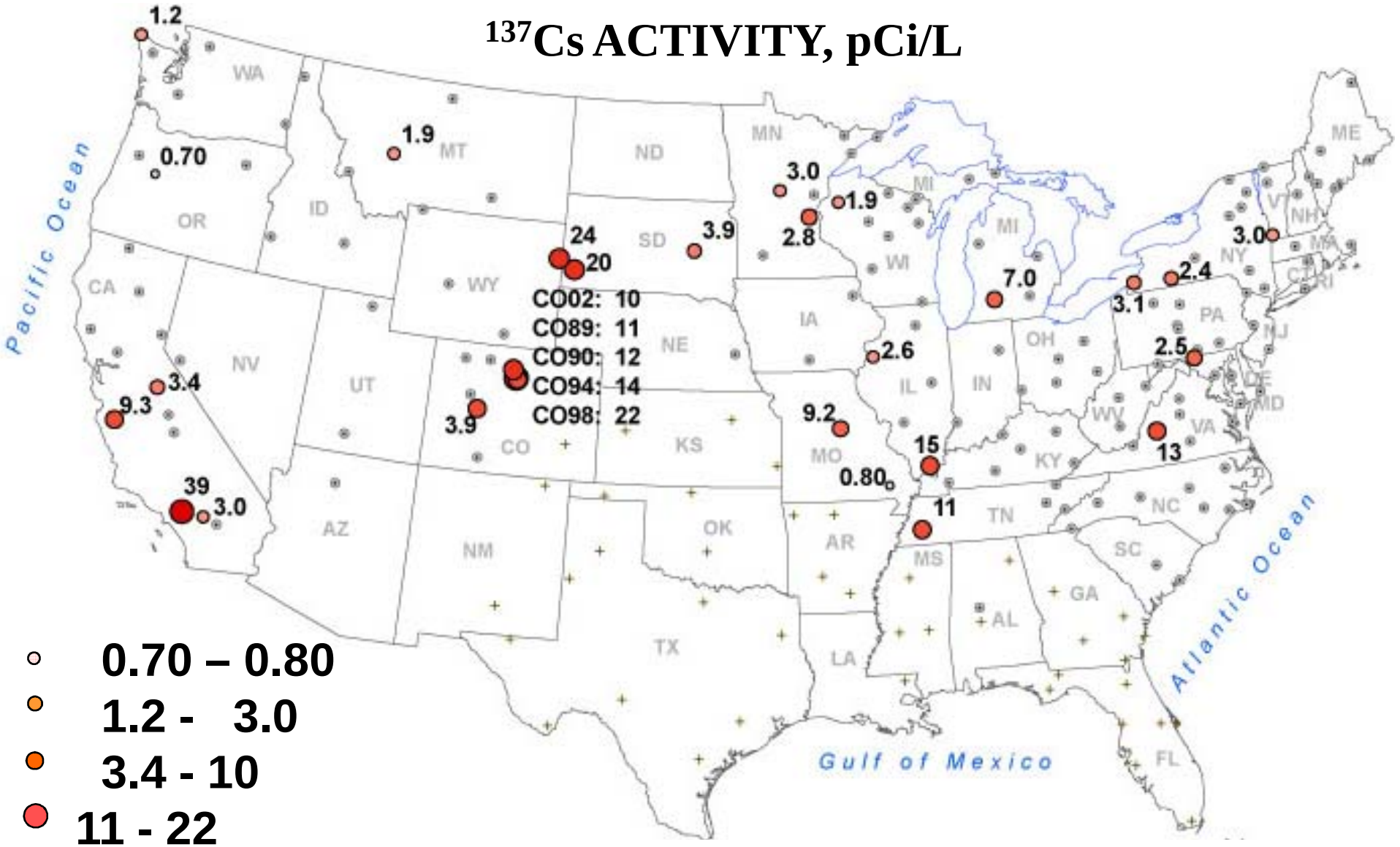
^{137}Cs ACTIVITY, pCi/L



- ○ 0.70 – 0.80
- ● 1.2 - 3.0
- ● 3.4 - 10
- ● 11 - 22
- ● 24 - 39
- \+ Plant Pathogen Study
- ○ Not detected

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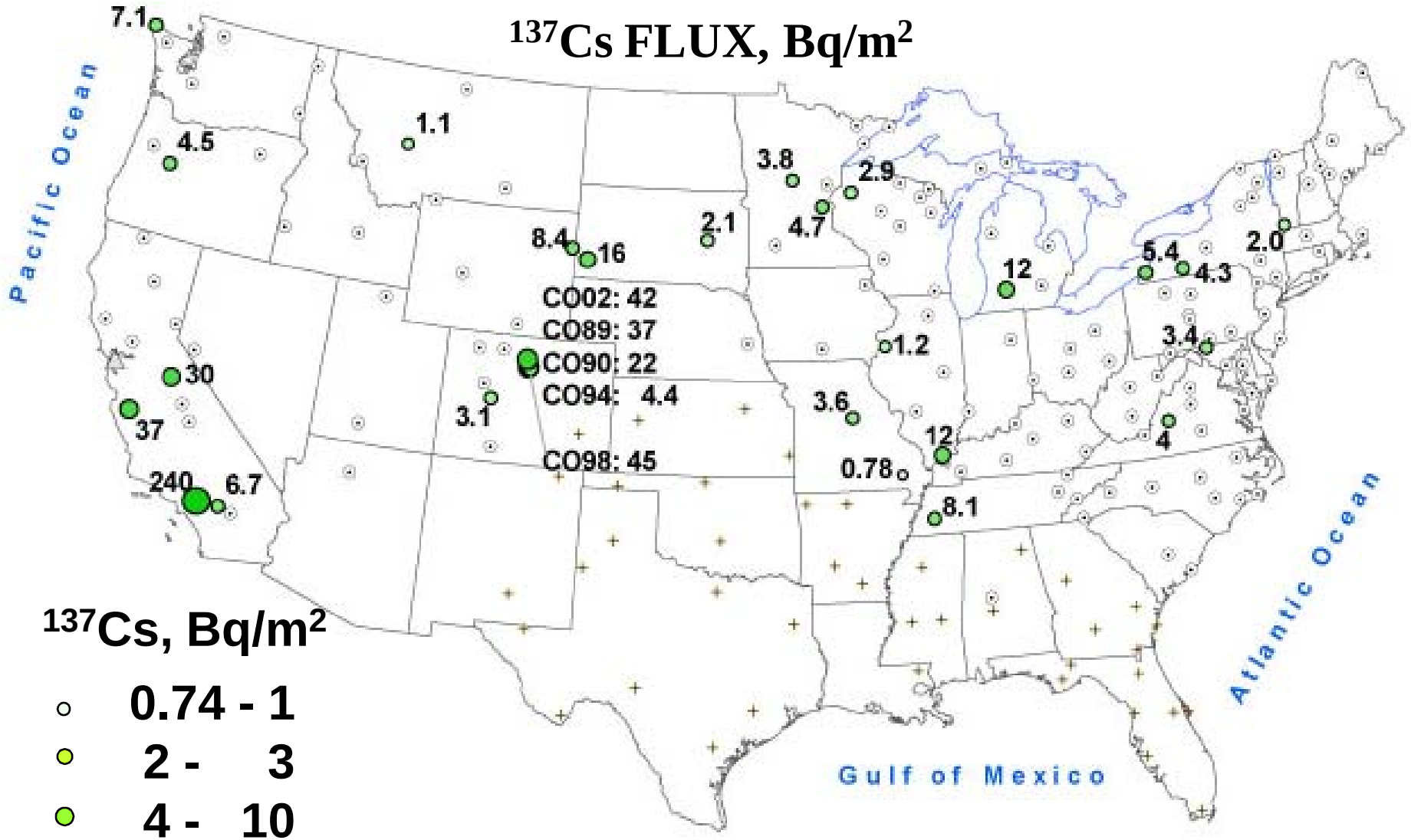
^{137}Cs FLUX, Bq/m^2



^{137}Cs, Bq/m^2

- ○ 0.74 - 1
- ● 2 - 3
- ● 4 - 10
- ● 20 - 40
- ● 74 - 240
- \+ Plant Pathogen Study
- ○ Not detected

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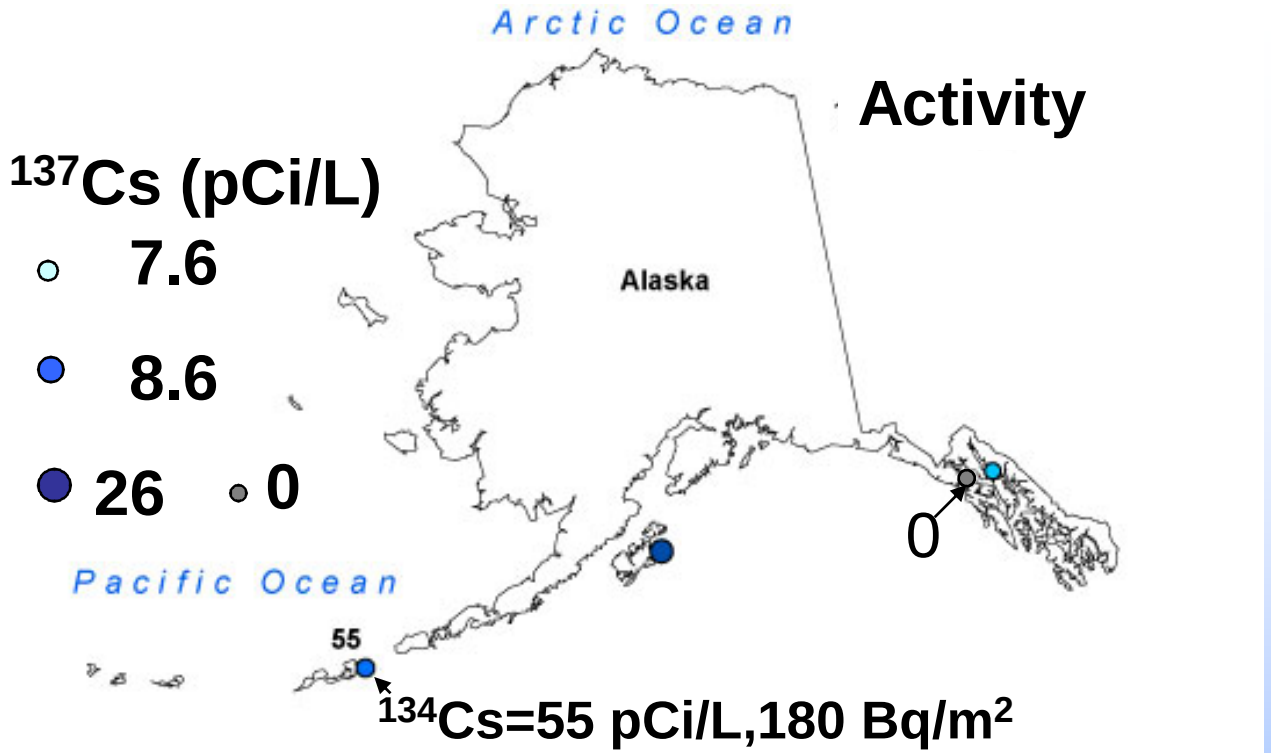
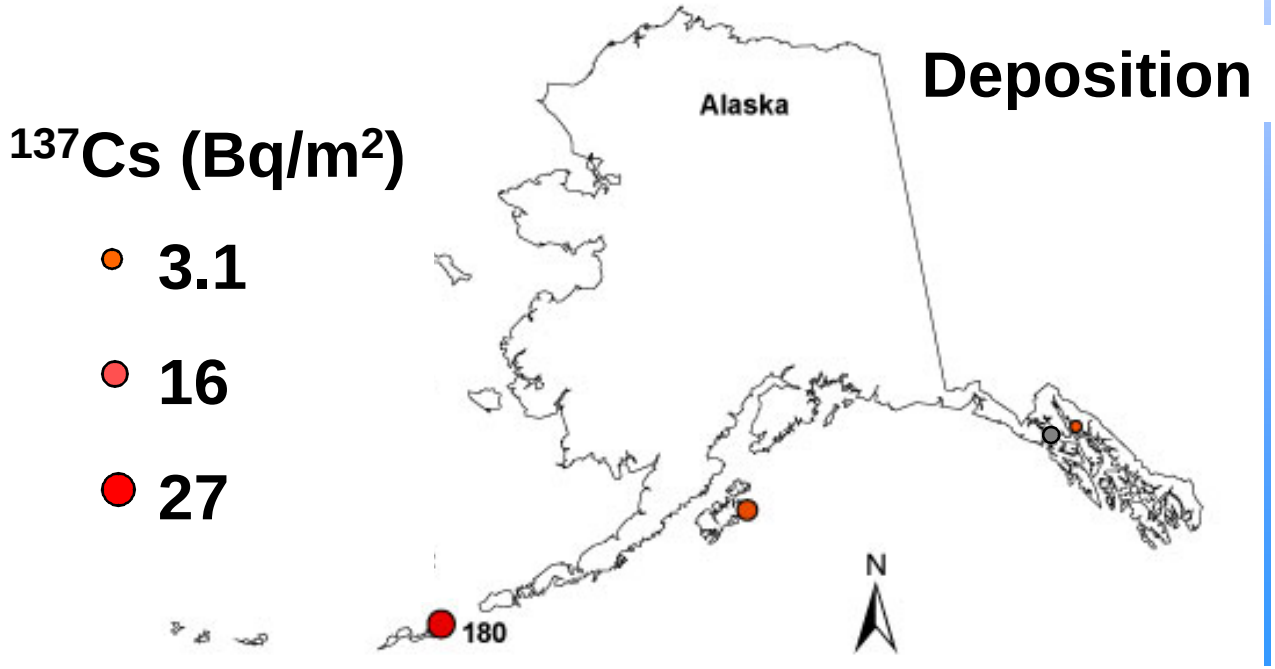
NADP IN ALASKA

^{134}Cs and ^{137}Cs





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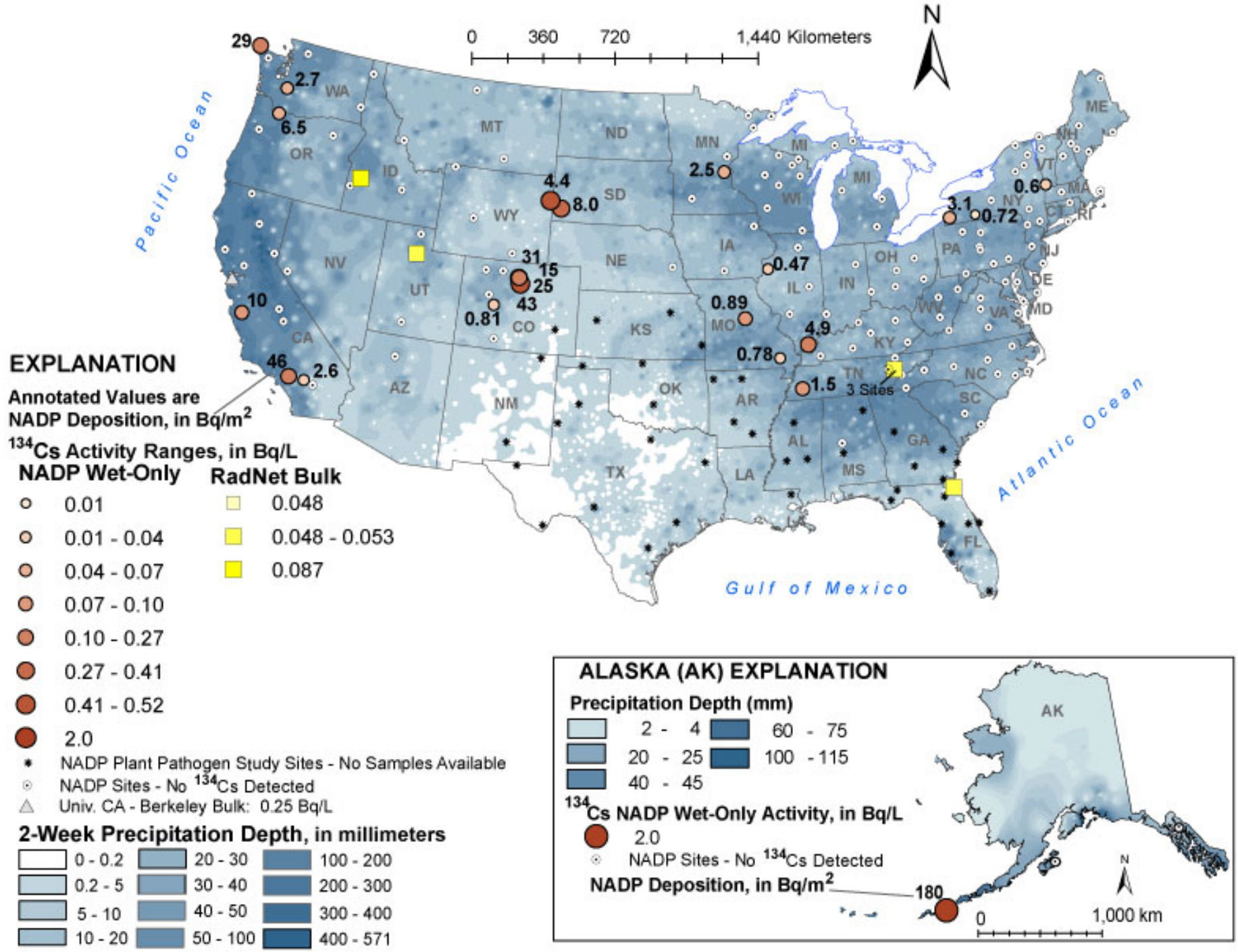
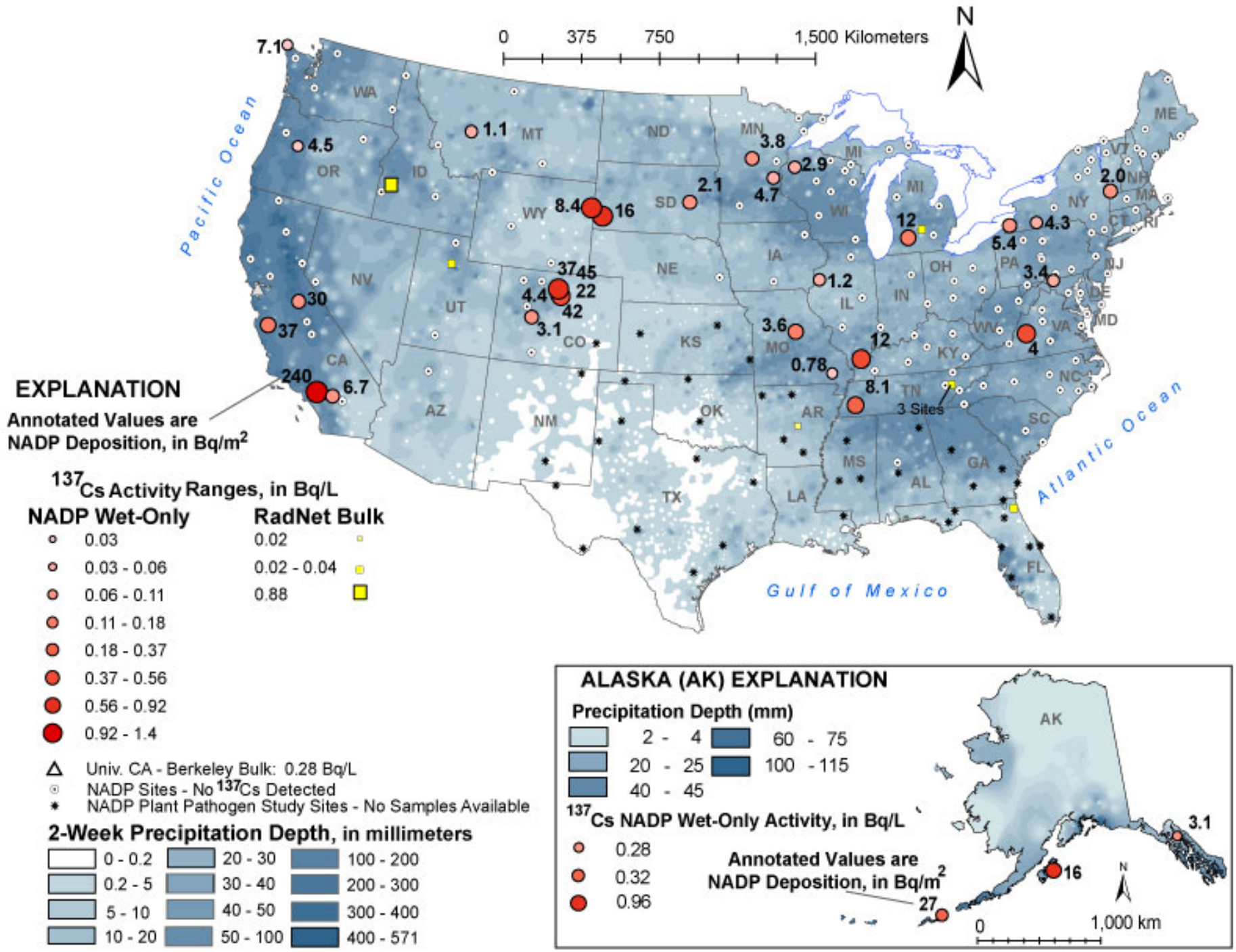
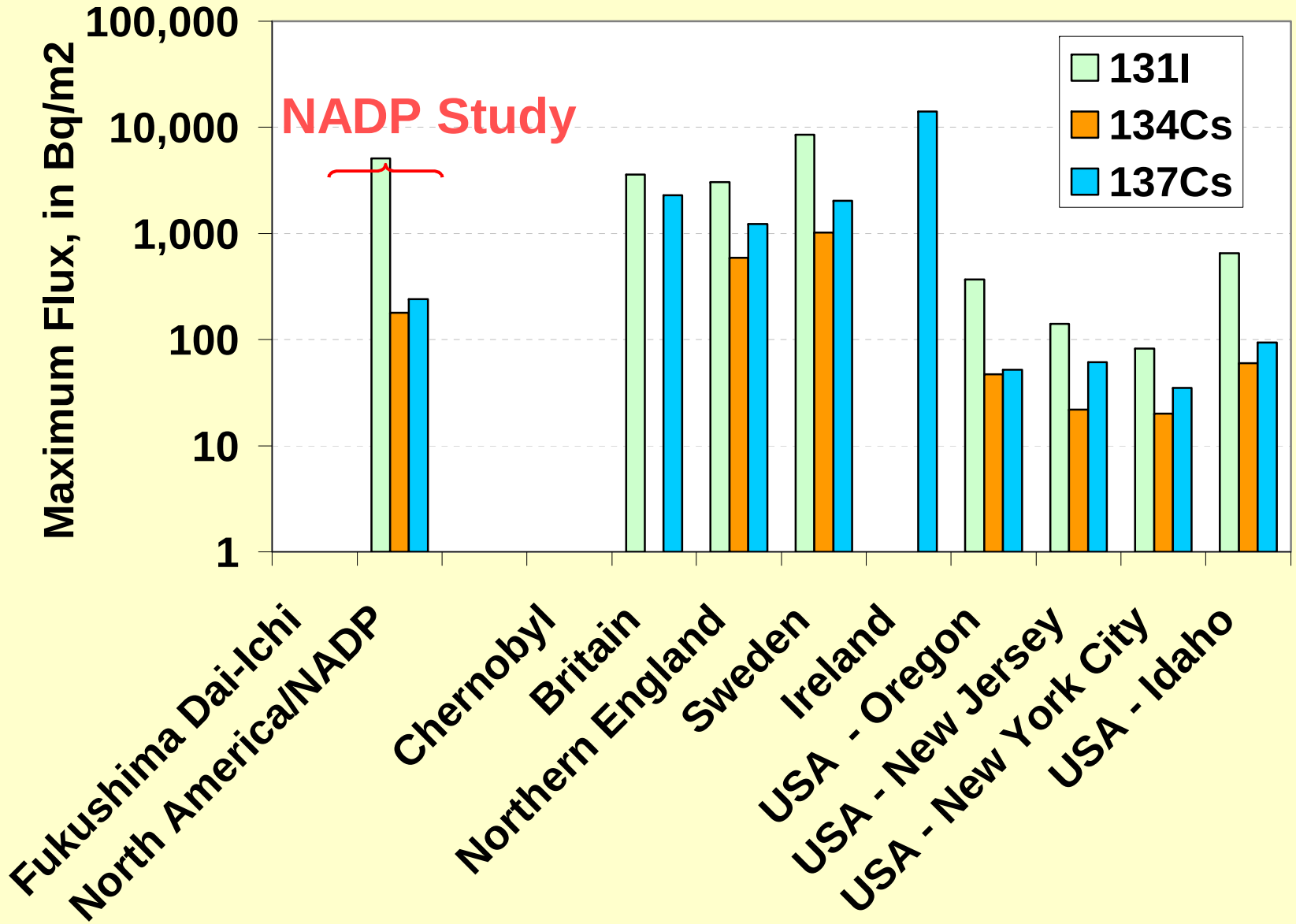

0 360 720 1,440 Kilometers
N
Pacific Ocean
Atlantic Ocean
Gulf of Mexico
WA
OR
CA
ID
NV
MT
WY
UT
AZ
CO
NM
ND
SD
NE
KS
OK
TX
MN
IA
MO
AR
LA
WI
IL
MS
MI
IN
KY
TN
AL
OH
WV
GA
FL
SC
NC
VA
PA
NY
ME
VT
NH
MA
CT
RI
NJ
DE
MD
29
2.7
6.5
4.4
8.0
2.5
31
15
25
43
0.81
10
46
2.6
0.89
0.47
4.9
0.78
1.5
3 Sites
3.1
0.72
0.6
EXPLANATION
Annotated Values are NADP Deposition, in Bq/m2
134Cs Activity Ranges, in Bq/L
NADP Wet-Only
0.01
0.01 - 0.04
0.04 - 0.07
0.07 - 0.10
0.10 - 0.27
0.27 - 0.41
0.41 - 0.52
2.0
RadNet Bulk
0.048
0.048 - 0.053
0.087
NADP Plant Pathogen Study Sites - No Samples Available
NADP Sites - No 134Cs Detected
Univ. CA - Berkeley Bulk: 0.25 Bq/L
2-Week Precipitation Depth, in millimeters
0 - 0.2
0.2 - 5
5 - 10
10 - 20
20 - 30
30 - 40
40 - 50
50 - 100
100 - 200
200 - 300
300 - 400
400 - 571
ALASKA (AK) EXPLANATION
Precipitation Depth (mm)
2 - 4
20 - 25
40 - 45
60 - 75
100 - 115
134Cs NADP Wet-Only Activity, in Bq/L
2.0
NADP Sites - No 134Cs Detected
NADP Deposition, in Bq/m2
180
AK
N
0 1,000 km


0 375 750 1,500 Kilometers
N
Pacific Ocean
Atlantic Ocean
Gulf of Mexico
7.1
4.5
1.1
8.4
16
2.1
3.8
2.9
4.7
12
5.4
4.3
2.0
3.4
1.2
3.6
12
0.78
8.1
4
3 Sites
37
45
22
42
4.4
3.1
30
37
240
6.7
WA
OR
CA
ID
NV
MT
WY
UT
AZ
CO
NM
ND
SD
NE
KS
OK
TX
MN
IA
MO
AR
LA
WI
MI
IL
IN
OH
KY
TN
MS
AL
GA
FL
SC
NC
VA
WV
PA
NY
ME
VT
NH
MA
CT
RI
NJ
DE
MD
EXPLANATION
Annotated Values are
NADP Deposition, in Bq/m2
137Cs Activity Ranges, in Bq/L
NADP Wet-Only
0.03
0.03 - 0.06
0.06 - 0.11
0.11 - 0.18
0.18 - 0.37
0.37 - 0.56
0.56 - 0.92
0.92 - 1.4
RadNet Bulk
0.02
0.02 - 0.04
0.88
Univ. CA - Berkeley Bulk: 0.28 Bq/L
NADP Sites - No 137Cs Detected
NADP Plant Pathogen Study Sites - No Samples Available
2-Week Precipitation Depth, in millimeters
0 - 0.2
0.2 - 5
5 - 10
10 - 20
20 - 30
30 - 40
40 - 50
50 - 100
100 - 200
200 - 300
300 - 400
400 - 571
ALASKA (AK) EXPLANATION
Precipitation Depth (mm)
2 - 4
20 - 25
40 - 45
60 - 75
100 - 115
137Cs NADP Wet-Only Activity, in Bq/L
0.28
0.32
0.96
Annotated Values are
NADP Deposition, in Bq/m2
27
16
3.1
AK
N
0
1,000 km

Fukushima vs Chernobyl



Wetherbee, G.A., Gay, D.A., Debey, T.M., Lehmann, C.M.B., and Nilles, M.A., 2012, Wet deposition of fission-product isotopes to North America from the Fukushima Dai-ichi Incident, March 2011, Table 1, Environmental Science and Technology, 46 (5), pp 2574–2582.

Context

- **Maximum ^{137}Cs deposition from Fukushima in USA would contribute approximately 3%-10% additional radioactivity to that present in a common square meter of soil (5 cm deep).**

- **Maximum NADP-measured ^{137}Cs wet deposition from Fukushima (240 Bq/m^2) is about 17% of the highest U.S. annual total wet-deposition estimated in New York, NY, and Birmingham, AL (1,400 Bq/m^2) during atmospheric nuclear testing in 1963.**

CONCLUSIONS

1. Detectable ^{131}I, ^{134}Cs, & ^{137}Cs
   20% of sampled locations.

2. Estimated Deposition (FLUX) Ranges:

^{131}I:	60. – 5,100 Bq/m^2	- 5 SITES
^{134}Cs:	0.47 – 46 Bq/m^2	- 25 SITES
^{137}Cs:	0.74 – 240 Bq/m^2	- 33 SITES

3. Spatial distribution of deposition and source region consistent with NOAA HYSPLIT back trajectory analysis.

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CONCLUSIONS

4. Fission products associated with particles < 0.45 mm, OR dissolved species.

5. NADP demonstrated a national capability to monitor unexpected releases of radionuclides to the environment.

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science for a changing world

LESSONS LEARNED

1. Prioritize whole water precipitation samples & archive filters for possible later analysis.

2. Improve interagency coordination for sample analysis, especially for short-lived isotopses such as ^{131}I.

Protocols to Consider

1. Dedicate a collector solely for the purpose of collecting wet deposition for radionuclide monitoring.

2. Acidify (preserve) samples in the bucket BEFORE transfer of sample to bottle.

3. Homogenize (“stir”) sample BEFORE transfer to bottle instead of decanting off liquid only to obtain particulates.

Future Work

1. Reanalysis of samples for ^{90}Sr.

   We Need a lab to do radiochemistry!

2. Proposed ^{3}H network for southeastern USA.

ACKNOWLEDGMENTS

RoseAnn Martin (USGS/BQS)

Brenda Riney (NADP/CAL)

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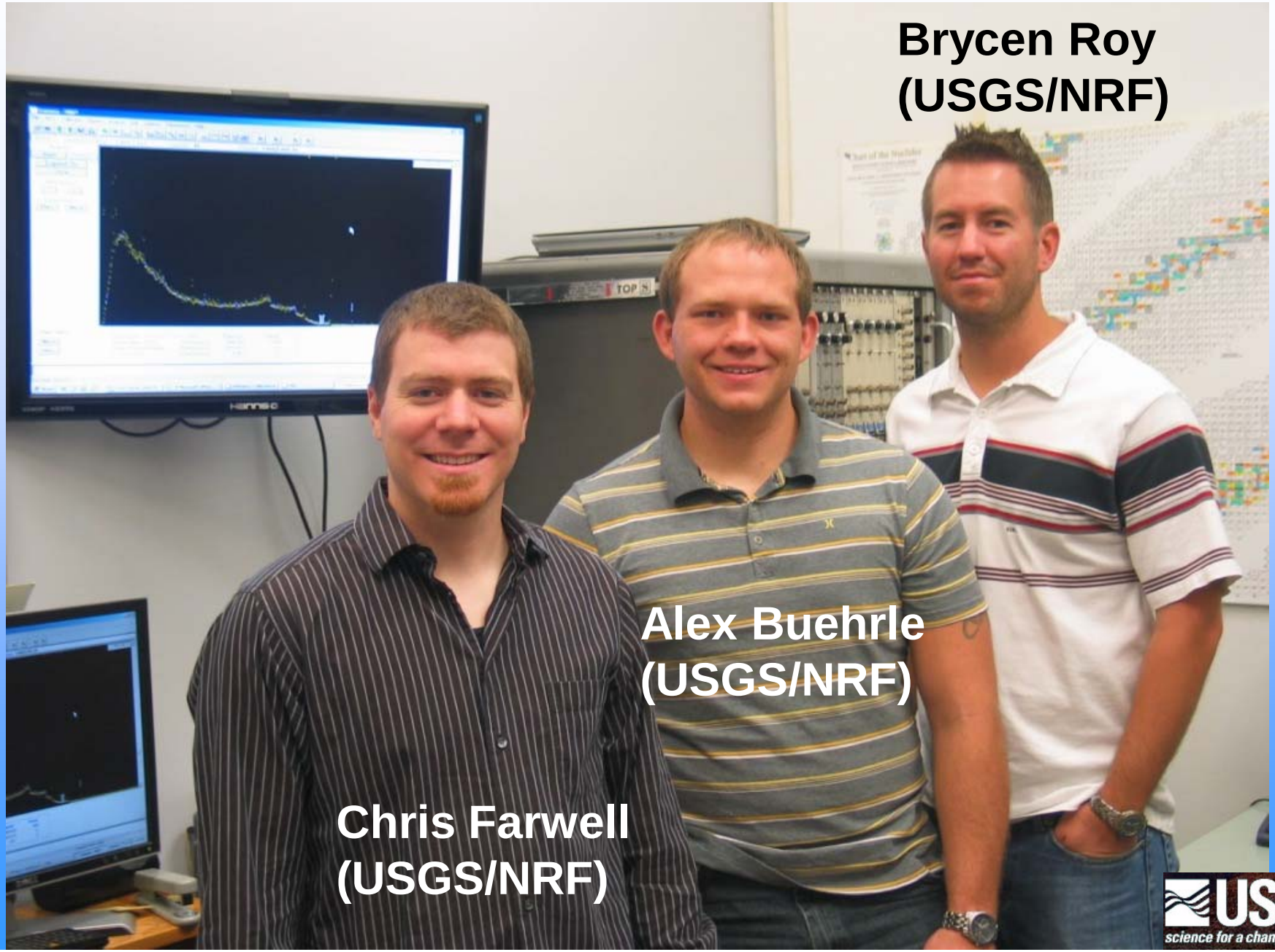
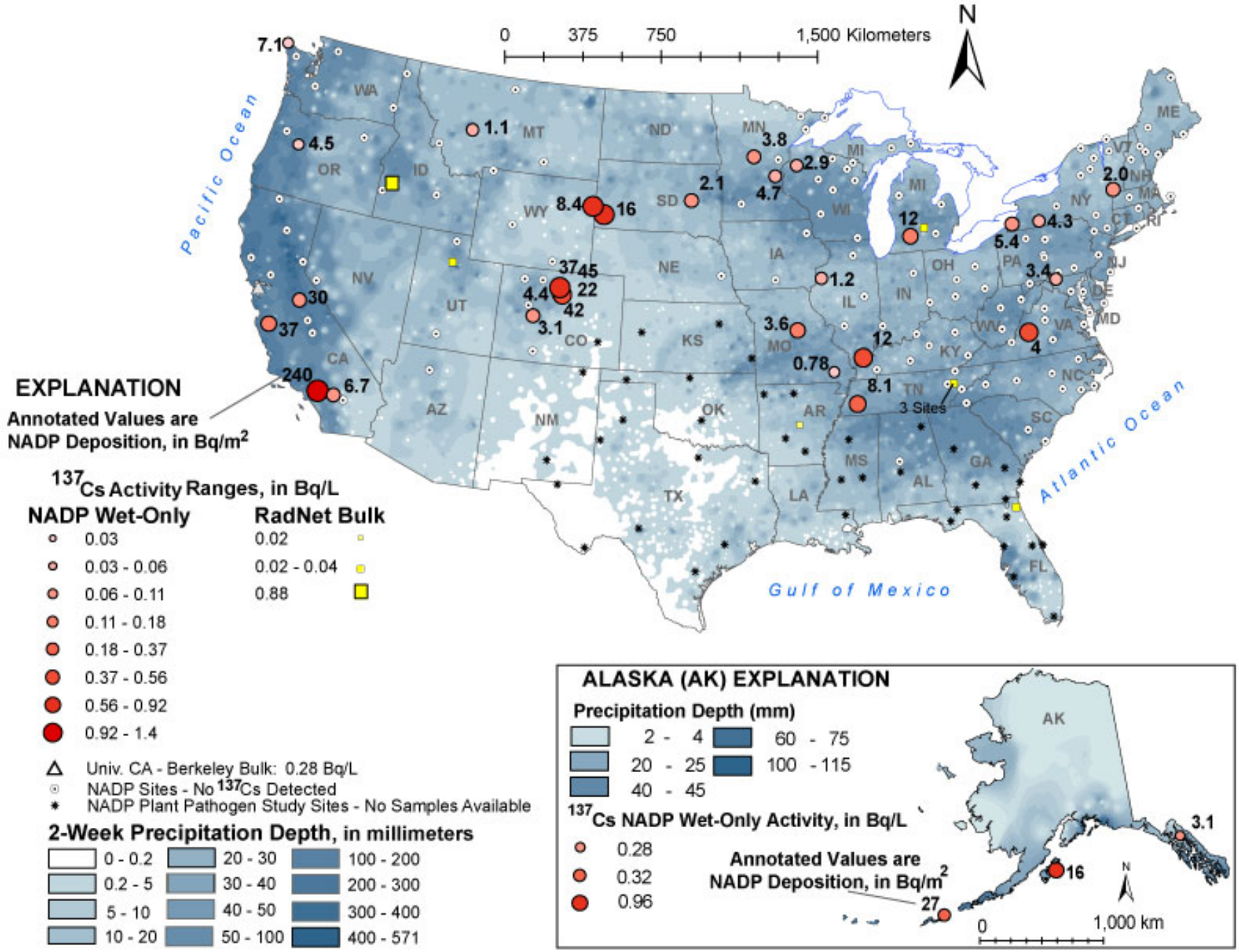
ACKNOWLEDGMENTS




0 375 750 1,500 Kilometers
N
Pacific Ocean
Atlantic Ocean
Gulf of Mexico
7.1
4.5
1.1
30
37
240
6.7
8.4
16
2.1
37
45
22
4.4
42
3.1
3.8
2.9
4.7
12
1.2
3.6
0.78
12
8.1
3 Sites
5.4
4.3
2.0
3.4
4
EXPLANATION
Annotated Values are
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137Cs Activity Ranges, in Bq/L
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0.03 - 0.06
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0.18 - 0.37
0.37 - 0.56
0.56 - 0.92
0.92 - 1.4
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0.02 - 0.04
0.88
Univ. CA - Berkeley Bulk: 0.28 Bq/L
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40 - 50
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100 - 200
200 - 300
300 - 400
400 - 571
ALASKA (AK) EXPLANATION
Precipitation Depth (mm)
2 - 4
20 - 25
40 - 45
60 - 75
100 - 115
137Cs NADP Wet-Only Activity, in Bq/L
0.28
0.32
0.96
Annotated Values are
NADP Deposition, in Bq/m2
27
16
3.1
0 1,000 km